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ORELA Nuclear Data Workshop

D. W. Nigg
J. D. Baker
C. A. McGrath
C. L. Riddle
A. Caffrey
J. Anderson
J. D. Cole
M. W. Drigert
R. Aryaeinejad
R. V. F. Janssens
B. J. Micklich
G. Ter-Akopian

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D.W. NIGG ,*J.D. BAKER, C.A. MCGRATH, C.L. RIDDLE, A. CAFFREY, J. ANDERSON*, J.D. COLE, M.W. DRIGERT, R. ARYAEINEJAD,
Idaho National Laboratory, Idaho Falls, ID, USA

R.V.F. JANSSENS, B.J. MICKLICH
Argonne National Laboratory, Chicago, IL, USA

G. TER-AKOPIAN
Joint Institute for Nuclear Research, Dubna, Russia

Introduction

Studies conducted in connection with the Department of Energy (DOE) Generation IV (Gen-IV) and Advanced Fuel Cycle (AFC) programs show that the transuranic nuclides can strongly influence the neutronic behavior of some advanced nuclear energy systems of interest. Essentially all integral nuclear parameters computed using modern reactor physics codes and data libraries are affected by propagation of uncertainty in the underlying nuclear data used in the computational models. These parameters include:

- Criticality (multiplication factor)
- Reactivity feedback coefficients (e.g., Doppler, Coolant Void)
- Kinetics parameters (e.g., Effective Delayed Neutron Fraction)
- Reactivity loss during irradiation (Burnup Swing)
- Peak power value
- Conversion ratio of sustainable cores
- Transmutation potential of burner cores
- Max dpa, maximum helium- and hydrogen-production, etc.
- Decay heat, radiotoxicity, and neutron and gamma radiation levels

However, the necessary cross section information may be unavailable with the required accuracy from the current nuclear databases for some key nuclides of interest. As a specific example, recent sensitivity analyses for the Very High Temperature Gen-IV reactor concept (Taiwo et al, 2005a, Salvatores et al., 2005, Taiwo et al, 2005b), which features a somewhat harder thermal neutron spectrum and a significantly higher fuel burnup target than is the case for standard light-water reactors, show a potential need for improved cross sections for some isotopes, including ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{243}Am, primarily in the resonance energy range, in order to satisfy the defined accuracy requirements on key computed integral parameters. Other transuranic nuclides may be of similar importance for some of the other Gen-IV concepts, although the primary needs appear to be currently focused around a few plutonium and americium isotopes. The target accuracies identified in these studies are very stringent and will be a challenge to achieve in many cases.

In this article we briefly review the conclusions and recommendations of recent international workshops on nuclear data needs for Gen-IV. Following this is a discussion of two specific activities undertaken by the Idaho National Laboratory (INL) to participate in the international effort to address these needs.

Nuclear Data Workshops

There have been three recent workshops on nuclear data needs for Gen-IV. The first was held at Brookhaven National Laboratory. The participants were primarily from US National Laboratories and Universities, with a few European participants. The second workshop was organized as an embedded meeting at the American Nuclear Society Topical Meeting on Reactor Physics held in Chicago during April 2004 (PHYSOR04). The third workshop was held in Antwerp, Belgium in April 2005 and was primarily organized by the European Institute for Reference Materials and Measurements in Geel, Belgium and was sponsored by the US Department of Energy, Atomic Energy of Canada, and Euratom. This latter workshop was a follow-on to the two previous workshops with broader international participation, including representatives from France, Germany, Belgium, Romania, Italy, Netherlands, Russia, USA, Canada, S. Korea, Finland, Czech Republic, Japan, Mexico and various international organizations including IRRM (Geel, Belgium), OECD-NEA, IAEA, etc. There were discussions of fuel development, intercomparison of evaluations, sensitivity studies, and basic discussions of nuclear data measurements – providing a broad perspective and context.

Key conclusions of the Brookhaven workshop included the following:

- High burnup operation of the VHTR might require re-evaluation of transuranic data (cross sections, decay data, and fission yields). Differential measurements may be needed for selected nuclides
- Fast spectrum systems (GFR, LFR, and SFR) to be used within a closed fuel cycle require additional evaluation of data for transuranic nuclides, particularly minor actinides, as well as integral measurements for validation of differential (basic) data and their processing tools
- Non-conventional structural, coolant or fuel-matrix materials may necessitate new evaluations or measurements of basic data
- A systematic approach based on sensitivity and uncertainty analysis is required for further specifying data needs. (This led to the sensitivity studies published in 2005)
- Currently available experimental facilities, equipment, accelerator targets, and personnel required to support necessary differential nuclear data measurements should be able to address the anticipated need for data. Key U.S. facilities identified include those at LANL, ORNL, RPI, and the INL experimental apparatus located at ANL/IPNS

- There should be a strong emphasis on maintenance of the relevant experimental capabilities and on development of a single national collaborative effort, coordinated with relevant international activities, that will provide the necessary information, with appropriate levels of validation, in a manner that makes best use of what will almost certainly be limited financial resources
- A coordinated mechanism should be developed to facilitate the acquisition, maintenance, storage, distribution, and community usage of sample targets, especially purified stable isotopes and actinides. An assessment of the nuclear materials available for this effort should be performed

Key conclusions of the Antwerp workshop were as follows:

- Data uncertainties and assessment of their impact are keys to improvement of reactor and fuel cycle codes. New approaches to measurement and evaluation will be important in reducing the current uncertainties
- In current fuel cycle scenarios uncertainties in ^{238}U, ^{239}Pu, some of the higher plutonium isotopes and possibly americium will be more important than the higher actinides (ANL and CEA Studies)
- Some attention to more accuracy in fission product yields appears to be needed
- Several non-fuel materials (Bi, Pb,C, Si, Zr) may need additional improvement.
- There is a clear need for improved covariance matrices to use in uncertainty studies

The INL Nuclear Data Initiative

The Idaho National Laboratory has made significant contributions to the international nuclear database over the years. Cross section measurements historically were performed at the Materials Test Reactor in the early days of the laboratory using various techniques. More recently, the INL has focused its efforts in nuclear physics on fundamental studies of the fission process using spontaneous fission sources as well as gated accelerator neutron sources with fissionable targets. The current nuclear data initiatives undertaken by the INL involve collaborations with Los Alamos National Laboratory (LANL) and, separately, with Argonne National Laboratory. The effort builds on historical INL capabilities not only in nuclear physics but also in radiochemistry.

The collaboration with Los Alamos involves production of actinide targets for use in capture and fission measurements performed by LANL at the Los Alamos Neutron Science Center (LANSCE). The current focus is on capture measurements at the LANSCE Lujan Center using the DANCE (Detector for Advanced Neutron Capture Experiments) detector array. DANCE, shown in Figure 1, is a 4π calorimeter with BaF scintillation detectors. Targets are currently prepared for use in DANCE using an electrodeposition process, although development of a direct metal vapor deposition process for this purpose is currently under development at INL. Figure 2 shows a ^{239}Pu target delivered to LANL by INL in June 2005. Future plans call for preparation of ^{242}Pu targets as well as additional targets for other actinide studies using the LANL apparatus.

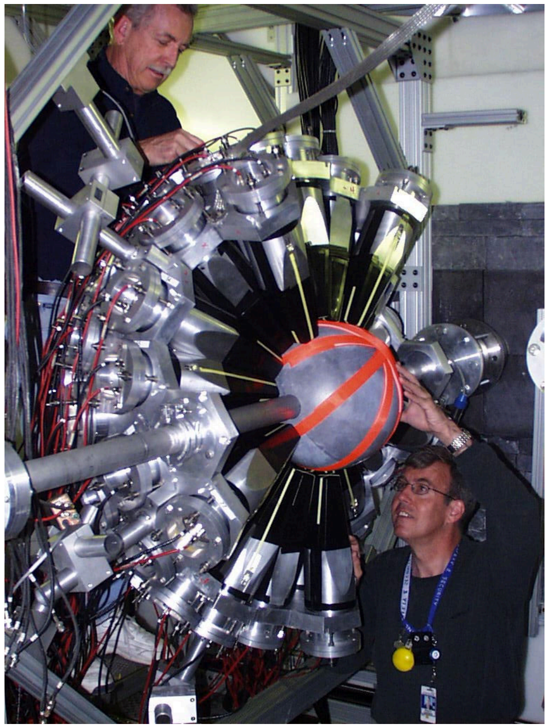

Figure 1. DANCE array under construction at LANL. Photo provided by J. Ullmann, LANL.

Figure 2. Plutonium-239 target prepared by INL for use in the LANL DANCE array.

The INL collaboration with ANL involves use of sophisticated detector arrays, supporting electronics, and data acquisition systems originally established by INL on a beamline of the Argonne Intense Pulsed Neutron Facility (IPNS) for use in fundamental studies of the fission process. The INL apparatus, which consists of an array of multiple types of multiple detectors operated in coincidence, with correlation of each observed event to the time of flight of the neutron that induced it, also offers an innovative method for cross section measurements as well. The immediate measurement goals involve measurement of neutron induced interaction cross sections for ^{240}Pu ,^{242}Pu, ^{41}Pu, and possibly ^{241}Am, with measurements for other nuclides of interest for advanced reactor physics applications to follow later.

The ANL/IPNS facility is a spallation neutron source with a moderated neutron beam that has a neutron spectrum at 12 and 20 meters as illustrated in Figure 3. The shown in Figure 3 are the results of average measured intensities and MCNP calculations performed at IPNS. For perspective, a direct comparison of flux intensity is shown between IPNS and the Los Alamos Lujan Center at lower neutron energies where explicit numbers, in similar units, are available, in particular for epithermal neutrons.

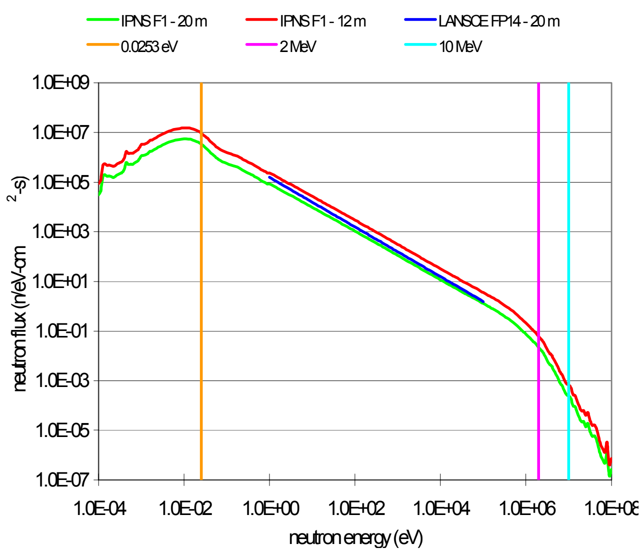

Figure 3. Plot of the neutron spectrum on the F3 beam line used for the INEEL apparatus for distances of 12 and 20 meters and the extrapolated values for FP14 at LANSCE. Energy markers are shown for the readers benefit.

The uncertainty of the energy of a neutron that induces an interaction of interest in the target is determined largely by the uncertainty on the n-TOF from the time width of the proton pulse. In addition to this important parameter, the pulse rate and the flight path length interact to limit the useable energy range of the neutrons. At IPNS the pulse rate is 30 Hz and the proton pulse full width is 70 ns. All beam lines at IPNS are heavily shielded and evacuated so that backgrounds are reduced. The low background at IPNS is an important factor for the long runs needed for experiments that achieve low statistical uncertainty.

The INL apparatus was originally installed at IPNS to perform experiments using induced fission of actinide targets for prompt information concerning fission yields by isotope pairs, nuclear structure information for prompt de-excitation of the fission products, multiplicity of both neutron and gamma rays by isotope pairs, and isotope pair distributions for fission cluster models. These efforts are extensions of spontaneous fission studies on ^{252}Cf and ^{242}Pu conducted with arrays of HPGe detectors at INL, Oak Ridge National Laboratory (ORNL), Lawrence Berkeley National Laboratory (LBNL), and finally on GAMMASPHERE at both ANL and LBNL. This work has produced over 100 publications on the nuclear structure of fission products prior to beta decay, fission yields by isotope pairs, and explicit neutron multiplicity as correlated to specific fission pairs.

Two years ago efforts began to modify the INL apparatus at IPNS to provide the capability to measure neutron interaction cross sections as a function of incident neutron energy, branching rations for the production of different isotopes by neutron capture or fission, and cross sections for the production of independent yields from actinide fission. The INL experimental technique allows a model-independent measurement of the neutron interaction cross section to be made over a continuous energy range from a few meV to above 2 MeV without breaking the measurement into different energy segments. With approximately 4000 hours of beam time available for measurements in one year, low statistical uncertainty can be achieved.

The INL detector array, shown in Figure 4, is composed of 12 Compton suppressed, high purity germanium (CSHPGe) detectors, eight fast neutron detectors (BC501 liquid scintillator), and a stack of up to 32 Silicon (Si) detectors interleaved with double-sided foils of actinide targets. The trigger electronics starts the digitization process if two of the CSHPGe, two neutron detectors, or a CSHPGe and a neutron detector produce a signal within a set coincidence time window. In this way three separate conditions can be used as independent triggers for determining a neutron interaction has occurred in a target. The coincidence is based on overlap timing with a time window of 50 to 100 nanoseconds.

The Si detectors are used to directly detect the fission fragments as these fragments recoil directly into the Si detectors. The Si detectors and the target foils are interleaved, each actinide foil has a selected thickness that will allow the low energy, light mass fission fragment to escape the target and enter the Si detector, which is in contact with the target material. A discriminator is used to reject α-particles and their pileup signal and accept only the fission fragment signal, which is a factor of ten greater in amplitude. These fission fragment signals in the Si detectors are used as a fourth trigger in the system. Si detectors are used instead of a fission chamber primarily because the rise time of the output pulse is faster by roughly a factor of ten. In addition, since the Si detector is in contact with the actinide target, gamma rays observed from the fragment have no Doppler shift or broadening due to emission in flight. There are other advantages with size, less support electronics, better α-particle discrimination, and low mass material. The energy output of each Si detector is also digitized and included in the data packet.

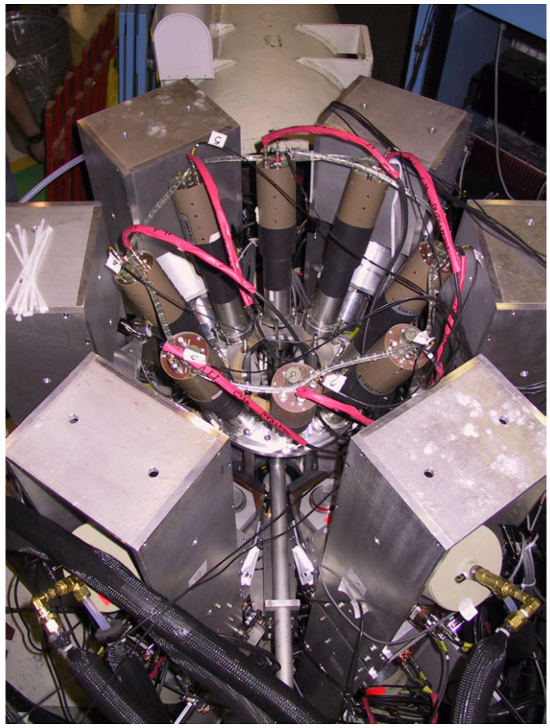

Figure 4. INL detector array for nuclear data measurements at ANL/IPNS.
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Signals from the four triggers are combined logically in various ways to produce an event signal. This event signal is used to trigger data acquisition of digitized detector signals, time relationships between the detector signals, and as a stop on a multi-stop time-digital converter (TDC). In this way a multi-parameter data packet is acquired for each observed radiation event and stored in list mode format. Since the system is configured to respond to coincidence events, and the prompt timing of an event is from 10^{-22} seconds to 10^{-12} seconds, a single trigger can result in multiple radiation types being included in the event. The simplest example is that of a fission fragment being detected in a Si detector and single gamma ray or neutron radiation also observed as will be shown later.

With the ability to select the events by post experiment sorting the multiplicities of both neutrons and gamma rays can be determined. The production rates of the selected gamma rays arising from the prompt fission fragments or the excited isotopes produced by neutron capture are used to determine the appropriate reaction cross sections as a function of the incident neutron energy, determined from the event-by-event neutron TOF parameter. In addition to information required to extract absolute reaction cross sections, the acquired data sets also will contain the information needed to extract independent fission fragment yields that can be used to validate accepted values in a model independent manner.

The most important aspect of this powerful ability to select events by sorting and then determine cross sections is to reduce background and events unassociated with the reaction channel of interest. This reduces the error on the cross section by allowing an event set to be selected that only contains events that are from that particular reaction channel. For example, problems associated with measuring the fission cross section for material with high spontaneous fission rates can be handled by sorting events based on different conditions. This method also reduces the total error whereas the traditional method of beam-on, beam-off does not remove the spontaneous fission events from the beam on data set. Selecting fission events by requiring a Si detector signal, and then sorting on gamma rays from different fission pairs will provide information on contributions for the two types of fission processes. The distribution of neutrons and thus associated fragments pairs are different for the spontaneous versus induced fission. This is caused by the differences in the excitation energy of the fissioning nucleus. In spontaneous fission the nucleus is in its ground state. For induced fission the nucleus will be at an excited state due to the energy brought in by the incident neutron and by the rearrangement of the population of the nuclear orbitals in the nuclear system after the neutron is captured.

The INL apparatus at IPNS thus has some key unique capabilities, supported by the IPNS facility itself. The most important is the ability to take coincidence data associated with a particular nuclear event as noted above. An array of detectors could be operated in this manner at other facilities as well, but at IPNS two important enabling features are available: 1) an intense flux of neutrons and, 2) the availability of the beam for long experimental measurements. These two facts allow low statistical uncertainties to be attained. Achieving a goal of $\sim 10^9$ events observed and stored by the data system requires over 100 days of beam time. This long experimental time is available at IPNS since the INL apparatus is on one beam line and not affecting experiments at other locations in the facility. The other unique capability associated with the INL protocol is related to the ability to perform nuclear event based data collection and post analysis by sorting data into subsets based on physics conditions. By imposing multiple conditions and sorts via software or computer processing in selecting data sets for detailed analysis, the "cross talk" between channels can be minimized in ways that are not possible in the hardware of the electronics. The simplest and easiest to understand is the Si detector trigger to separate fission events and all other events. The non-fission events can be further sorted look at other reaction channels. This capability has produced exceptional results in nuclear structure and spontaneous fission studies and is also applicable to the problems of measuring various neutron cross sections of actinide isotopes.

The post analysis capability based on data selection of reaction channels provides results that are self-consistent across the larger experimental data set. This means that the ability to use different approaches in sorting can provide results that provide consistency checks that are otherwise not available. An example of this is the case of the determination of a fission cross section by direct selection of the observation from the Si detectors and the cross section determined from sorting on gamma rays from the highest yield fission fragments. Although the statistics in these two subsets of data will be different, the cross section should have the same result in both cases. This is a powerful tool to check the results and provide a consistency not in previous work.

Targets used by INL at the IPN facility are fabricated in Russia by collaborators at the Joint Institute for Nuclear Research (JINR). The targets are metal foils, not oxides, on an appropriate backing and of the thickness needed to allow the light fission fragments to escape the target and enter the silicon detector that is in contact with the target material.

The removes the need for large corrections of the incident neutron flux that must often be done in the case of oxide targets. In addition, the vapor deposition of metal onto metal backing gives excellent stability to the targets and reduces the risk of contamination due to targets coming off the backing. The isotopic purity of the targets is greater than 98% for the principal isotope and a detailed chemical analysis is provide for each target batch and individual target characteristics such as mass per unit area, total mass, and other are provided.

Authorization was received in January 2005 for internal INL funding to perform a proof of principle experiment for cross section measurements at IPNS using ^{239}Pu standards. This experiment was recommended in a September 2004 international peer review of the INL/ANL proposal to perform some types of nuclear data measurements pertinent to the VHTR/Gen-4 program at IPNS. The experiment was successfully initiated in early May 2005 and data collection for the most recent accelerator operation cycle continued until the end of the cycle in late June. Significant results were obtained and presented at the DOE AFCI/Gen-IV Physics Working Group meeting at ANL July 19-20. Some key initial results are summarized below.

Figure 5 shows some initial time-of-flight (TOF) correlated fission event spectrum data from the INL silicon detectors over the energy range of interest from thermal to about 1 MeV. The prominent first resonance in plutonium at 0.3 eV is labeled, as are a few of the higher resonances. Recall that as one goes to the left in this plot the incident neutron energy increases to first order as the inverse square of the time of flight.

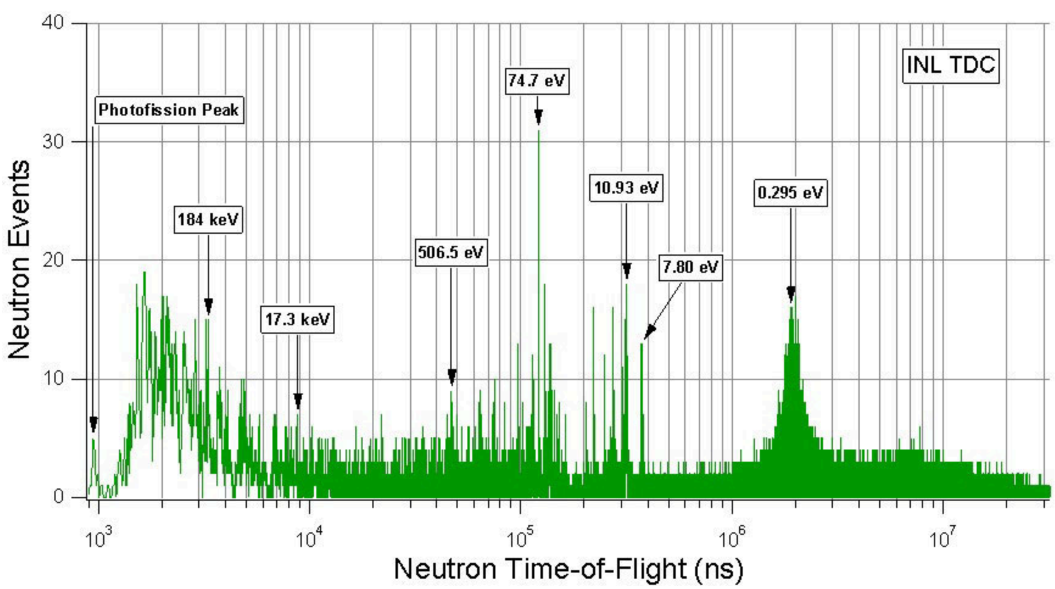

Figure 5. Initial ^{239}Pu Fission event spectrum from the IPNS nuclear data proof of principal experiment at ANL/IPNS. The neutron energy range spans 8 decades.

Figure 6 shows the same data with the energy range up to about 500 eV expanded to show more detail of the measured data for the prominent resonances.

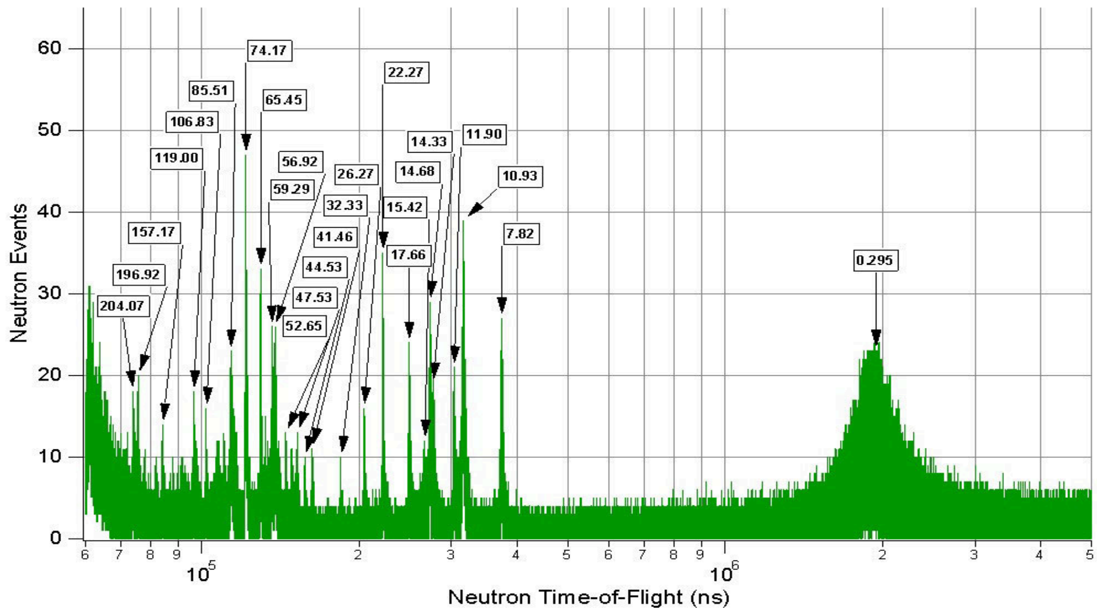

Figure 6. Fission event spectrum for ^{239}Pu. Expanded energy scale.

Figure 7 shows a further expansion of the energy scale to show detail in the energy range around 200 eV. The measured data are given in the lower plot, while the ENDF evaluation of the corresponding cross section for the same energy range is given in the top plot for comparison. Note the remarkable fidelity of the measured data relative to the standard evaluation. The statistical uncertainty on the measurements in these preliminary results is on the order of 10% at the resolution shown. However, these early demonstration experiments only involve collection of data over a few days of run time. In a precision calibration, or for a measurement of the cross section for an unknown sample, the run time would be much longer, to permit greater statistical accuracy on the order of the requirements identified by the various uncertainty propagation studies.

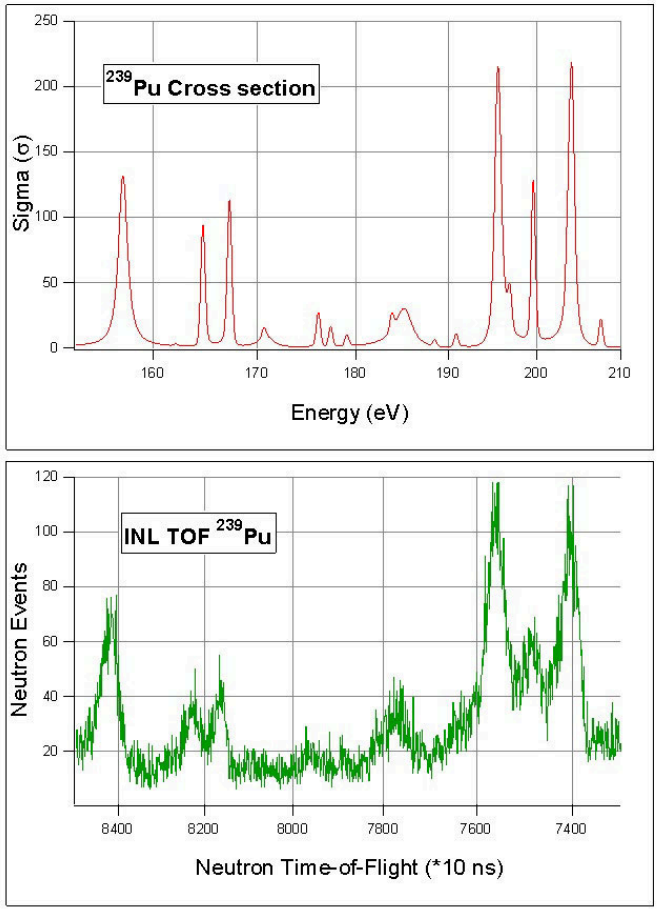

Figure 7. Comparison of the ENDF fission cross section (top) with the INL measured fission event spectrum.

Finally, and most significantly, it should be noted that the results given in Figures 5-7 are for "singles" data, i.e. data from the silicon fission detectors alone, with no coincidence gating. Figure 8 provides a striking initial example of what can be achieved by coincidence gating. In this case the signals from the gamma detectors in the INL/IPNS detector array are gated (by post-experiment sorting) in anticoincidence with the signals from the silicon fission detectors. This offers a self-consistent way of separating neutron capture events from fission events in a way that avoids the need to do individual experiments for each, or to normalize data to computational models. That is, an event detected by the gamma detectors is only "counted' if a fission is NOT detected at the same time. Figure 8 shows the results of this procedure for the energy range around 10 eV. The top plot once again shows the ENDF cross section (the standard for comparison) and the bottom plot shows the gated data, i.e. the neutron capture event spectrum (proportional to the capture cross section), separated from the fission event spectrum solely by experiment, in the same experiment - a key feature of the coincidence approach used at IPNS.

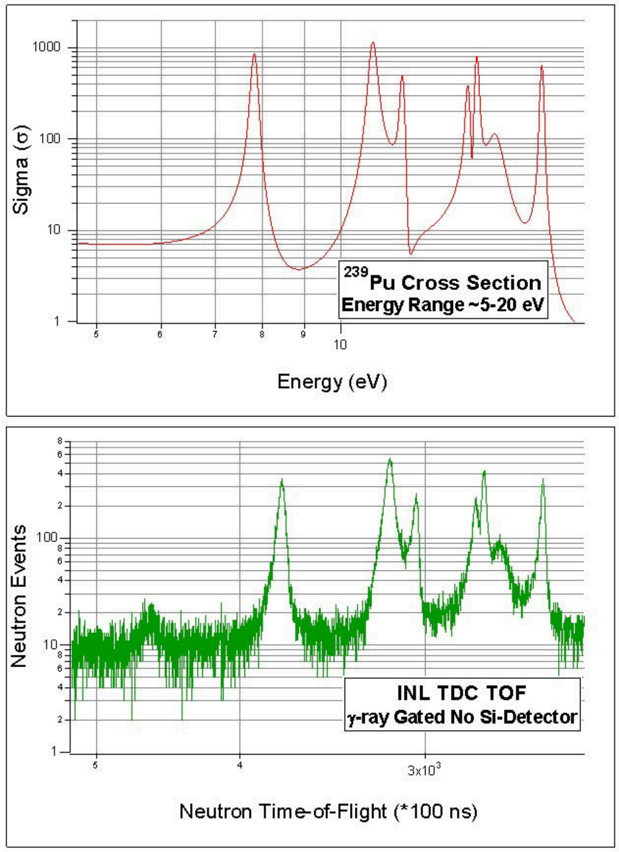

Figure 8. Capture event spectrum (bottom) for ^{239}Pu measured by INL using anticoincidence with fission, compared with the ENDF capture cross section evaluation (top).

Once again the high fidelity of the measured data is apparent. Furthermore, the coincidence approach also allows many other types of signals to be separated. For example, separating a neutron- induced event spectrum (i.e. cross section) from the very high background radioactivity present in some actinide targets of interest is very effectively enabled by this technique. Compensation for unwanted background due to target contamination is also possible. It may also be possible to resolve resonances in a manner that is not limited by the proton pulse width of the accelerator by capturing, through coincidence techniques, differences that may exist between the decay schemes of each resonance, although this remains to be demonstrated. Taken together, these features of the experimental apparatus and protocol offer the potential for improved accuracy in differential actinide cross section measurements important to VHTR as well as to other Gen-IV concepts.

References

T. A. Taiwo and H. S. Khalil (2005a) "Nuclear Data Needs for Generation IV Nuclear Energy Systems – Summary of U.S. Workshop", International Workshop on Nuclear Data Needs for Generation IV Nuclear Energy Systems, Antwerp, Belgium, April 5-7, 2005.

G. Aliberti, G. Palmiotti, M. Salvatores, T. K. Kim, T. A. Taiwo, I.Kodeli, E. Sartori, J.C. Bosq, J. Tommasi (2005) Sensitivity of Advanced Reactor and Fuel Cycle Performance Parameters to Nuclear Data Uncertainties", International Workshop on Nuclear Data Needs for Generation IV Nuclear Energy Systems, Antwerp, Belgium, April 5-7, 2005.

T.A. Taiwo, G. Palmiotti, G. Aliberti, M. Salvatores, T.K. Kim (2005b) "Uncertainty and Target Accuracy Studies for the Very High Temperature Reactor (VHTR) Physics Parameters", ANL-Gen-IV-051, August, 2005.